Flux gain for next-generation neutron-scattering instruments resulting from improved supermirror performance

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ABSTRACT

Next-generation spallation neutron source facilities will offer instruments with unprecedented capabilities through simultaneous enhancement of source power and usage of advanced optical components. The Spallation Neutron Source (SNS), already under construction at Oak Ridge National Laboratory and scheduled to be completed by 2006, will provide greater than an order of magnitude more effective source flux than current state-of-the-art facilities, including the most advanced research reactors. An additional order of magnitude gain is expected through the use of new optical devices and instrumentation concepts. Many instrument designs require supermirror (SM) neutron guides with very high critical angles for total reflection. In this contribution, we will discuss how the performance of modern neutron scattering instruments depends on the efficiency of these supermirrors. We outline ideas for enhancing the performance of the SM coatings, particularly for improving the reflectivity at the position of the critical wave vector transfer. A simulation program has been developed which allows different approaches for SM designs to be studied. Possible instrument performance gains are calculated for the example of the SNS reflectometer.

Keywords: Neutron scattering, neutron guides, supermirrors, instrument performance

1. INTRODUCTION

It is well known that neutron scattering is a powerful tool for the study of condensed matter because the wavelengths and energies of thermal and cold neutrons match well to the length and energy scales of solids and liquids.[1] The applicability of neutron scattering techniques, however, is limited by the relatively low flux of useful neutrons generated by today's research reactors or pulsed spallation sources, which is many orders of magnitude smaller than the flux of X-rays produced by contemporary photon sources. Recently, large efforts have been made to optimize existing and to develop new more powerful sources. The Spallation Neutron Source (SNS), which is already under construction at Oak Ridge National Laboratory, will become operational in 2006, and generate an effective neutron flux about one order of magnitude higher than the best existing neutron sources. Other approaches to gain intensity concern optimization of neutron optical components, development of new optical devices, and implementation of advanced instrument designs.[2] Simulation calculations indicate that these approaches should further increase the flux by up to one order of magnitude for particular SNS scattering instruments. Thus, the total intensity gain for SNS instruments can be as high as about two orders of magnitude, which will definitely move the quality of neutron scattering studies to new levels.

Supermirrors play an important role in most instrument designs at SNS. In this contribution, we will give a short overview about requirements on supermirrors and their current limitations. In particular, we have theoretically analyzed the intensity gain that may be achievable for the SNS Magnetism Reflectometer by increasing the performance of its supermirror guides. Our study is motivated by the fact that the experimentally measured reflectivity of supermirrors with m>2 differs significantly from theoretical predictions, which will definitely cause serious intensity losses after multiple reflections of the neutrons in the guide systems.

2. NEUTRON GUIDES AND GUIDE COATINGS

After neutrons have been produced in the source and moderated to useful energies, they need to be delivered to a variety of instruments, typically over a distance r, of some tens of meters. In the simplest case, an evacuated tube could be employed for transporting the neutron beam. This approach, however, would deliver only those neutrons to the sample with direct lineof-sight paths, resulting in very little neutron flux at the sample position (the neutron flux decreases with $1/r^2$). By using "neutron guides", however, much higher flux on the sample can be achieved.[3] Neutron guides typically consist of rectangular glass tubes internally coated with thin metal films, and neutrons are transport by reflection on these inner wall coatings. The gained flux consists of distribution of neutrons which have a higher degree of divergence compared to those having "natural divergence" (neutrons that would reach the sample if no guide were present). In the beginning, natural Ni, the element having the largest angle for total reflection for a given neutron wavelength, was employed as coating. Nowadays, more sophisticated "supermirror" coatings are used, as will be discussed below. The choice for a particular coating and a particular geometry for the guide system strongly depend on the requirements of the specific instrument that is fed by the guide and, of course, by financial constraints. In some cases high performance supermirrors are required, while in others Ni coatings are sufficient. At a spallation neutron source, almost every instrument occupies an "end position" on a guide; therefore, many guide designs include a funnel section in front of the sample in order to spatially compress the beam and enhance the neutron flux per sample area.

The angle of incidence, at which total reflection occurs, is called the critical angle ϑ_c. This angle is determined by the refractive index, which depends on neutron wavelength and on scattering length density of the reflecting material. For any given material, ϑ_c can be calculated as

$$\vartheta_c(\lambda)=\sqrt{2[1-n(\lambda)]}=\sqrt{\frac{N\cdot b}{\pi}}\cdot\lambda. \quad (1)$$

where n is the refractive index, λ the neutron wavelength, and $N\cdot b$ the product of number density N [atoms per unit volume], and scattering length b of the material. $N\cdot b$ is usually referred to as scattering length density. Since TOF instrumentation typically involves a large range of neutron wavelength, it is more appropriate to convert the critical angle into the corresponding critical momentum transfer

$$q_c=\frac{4\cdot\pi\cdot\sin\theta_c}{\lambda}=4\sqrt{N\cdot b\cdot\pi}. \quad (2)$$

If the interior of the guide is coated with pure Ni, all neutrons hitting its surfaces at angles lower than the critical angle of Ni (ϑ_c^{Ni}/λ = 1.7 mrad/Å) will be totally reflected. Such a neutron guide coating is usually defined as "m=1 mirror". In order to increase the critical angle of a coating, resulting in higher guide transmission, the reflecting Ni layer should be substituted by so-called "supermirrors". Supermirror-coatings consist of multilayers composed of thin films of materials showing large contrast in scattering length density, for example Ni and Ti.[4] The performance of a supermirror (SM) is described by the increase of its q_c-value compared to natural Ni:

$$q_c^{SM}=m\times q_c^{Ni}. \quad (3)$$

Neutron guides in pulsed facilities often differ from their reactor-based counterparts due to time-of-flight (TOF) based instrument operation. At reactors, long guide systems are employed for cold neutron research, and generally several instruments are fed by a single guide. In contrast, most SNS instruments that are currently being designed occupy single beam ports. Many instruments, such as reflectometers and small-angle scattering machines, need a relatively wide neutron bandwidth for their operation. Because of this, it is advantageous to build these instruments relatively short, typically 15-20 m. Key optical components of these instruments are channel beam deflectors ("beam benders") and focusing guides. The compact design of these devices requires supermirrors with very high critical angles for total reflection.

3. THEORETICAL ALGORITHMS FOR SUPERMIRROR DESIGNS

Mezei gave the first design "recipe" for artificially increasing the total reflection region of a neutron mirror beyond the critical momentum transfer of Ni. His approach is based on the idea of a continuously depth-graded multilayer, which he named "supermirror".[5,6] The working principle is based on Bragg reflections of neutrons by a system of double layers with varying periodicity. It is most effective if materials are used which have as large as possible scattering contrast, i.e. Ni and Ti. The bilayer period has to be changed slowly enough such that at any momentum transfer below the critical q of the supermirror, a sufficiently large number of bilayers scatter the neutron waves "in phase", i.e. to within ± 45° phase difference, to result in almost total reflectivity. Since the multilayer is usually covered by a Ni capping layer (typically several 100 Å thick), the supermirror reflective effect needs only exist for $q > q_c^{Ni}$. In the limit of very large q_c values, corresponding to small bilayer periodicity, the individual single layer thicknesses must be equal for optimal supermirror performance. In this "continuum" regime, refraction effects can be neglected. However, in the large bilayer thickness limit, i.e. close to the critical q of Ni, refraction effects play a significant role, demanding a correction of individual layer thicknesses.[7,8] Mezei's derivation of the supermirror layer sequence is based only on the real part of the materials optical index. Extinction effects, however, influence the maximum achievable reflectivities, particularly for high-m supermirrors.

A more sophisticated algorithm developed by Hayter and Mook takes into account the discrete nature of the layers.[8] It is based on a determination of the contribution of a given bilayer to the overall reflectivity in a sequence of layers. This method can easily take extinction into account. To construct a supermirror stack, the thicknesses of successive bilayers are chosen such that their reflectivity profiles intersect at half height. The starting point is defined by the intersection of the profile of the thickest bilayer and the critical edge of the substrate or an additional capping layer. This approach allows predetermination of a "design reflectivity" function for the supermirror. Usually this function is chosen such that the reflectivity declines linearly from practically unity at q_c of Ni to the desired reflectivity at q_c of the supermirror. For a given number of bilayers in the supermirror structure, varying the design reflectivity function allows for either optimizing reflectivity over a correspondingly smaller q-range or increasing the m-value of the supermirror at the expense of reflectivity.

Figure 1 illustrates the design of a supermirror. It shows how the neutron reflectivity of a Ni/Ti supermirror changes after sequentially increasing the numbers of bilayers. Film deposition usually starts with the thinnest layers on the well-polished glass or Si substrate (see upper part of Fig. 1) since the reflecting properties of those layers are most affected by roughness. In this example, an m=2 supermirror is calculated using Hayter and Mook's formula. In this case, 41 bilayers are required to achieve total reflection up to two times the critical edge of natural Ni (see lower part of Fig. 1). The first bilayer consists of 86.2 Å Ni / 72.1 Å Ti , whereas the 41st bilayer consists of 351.7 Å Ni / 123.2 Å Ti. Note that an extra 700 Å thick Ni capping layer is required to eliminate the reflectivity gap between of the critical edge of the substrate and the onset of the supermirror reflectivity at $q_c = 0.022 Å^{-1}$. The individual reflectivity curves have been calculated using a simulation program based on the well-known Parratt-formalism.[9] For simplicity, the effect of interface roughness has not been included in these calculations. This issue will be discussed separately in paragraph 4.3.

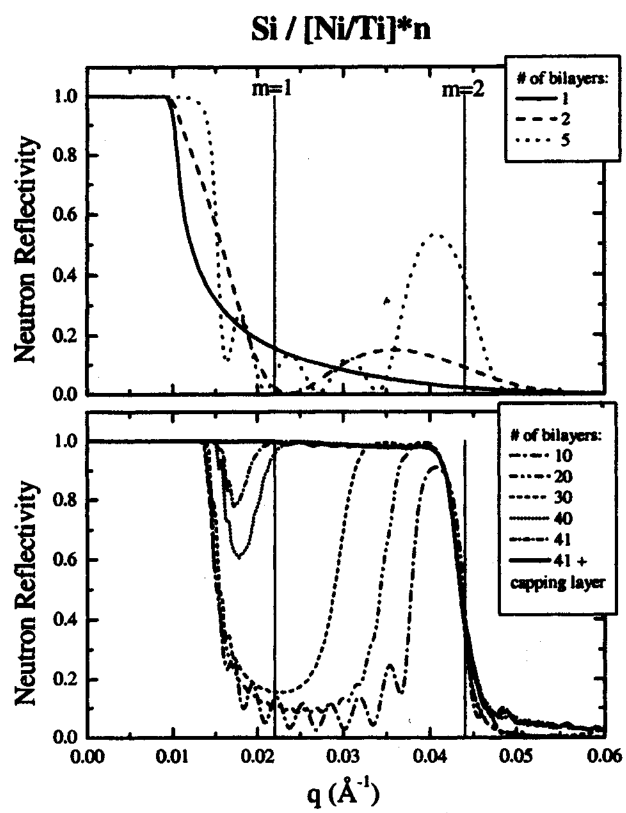

Fig. 1. Calculated reflectivity functions of an m=2 Ni/Ti supermirror after deposition of various numbers of bilayers.

4. SUPERMIRROR PERFORMANCE

In this chapter we will discuss parameters affecting the performance of supermirrors on the basis of Ni/Ti multilayers, in particular the number of bilayers, possibilities for isotope substitutions, interfacial roughness and other imperfections.

4.1 Number of bilayers

Figure 2 illustrates that, for a given design reflectivity function, the number of bilayers basically defines the q_c-value of the supermirror. It shows calculated neutron reflectivity curves of bulk Ni, and Ni/Ti supermirrors with increasing number of bilayers (see label). The layer sequences were calculated using the Hayter and Mook approach. It can be seen in Fig. 3 (derived from results shown in Fig. 2) that the increase in q_c is non-linearly related to the number of bilayers. High-m values require increasingly larger numbers of bilayers. To achieve m=3, for example, 250 bilayers are needed.

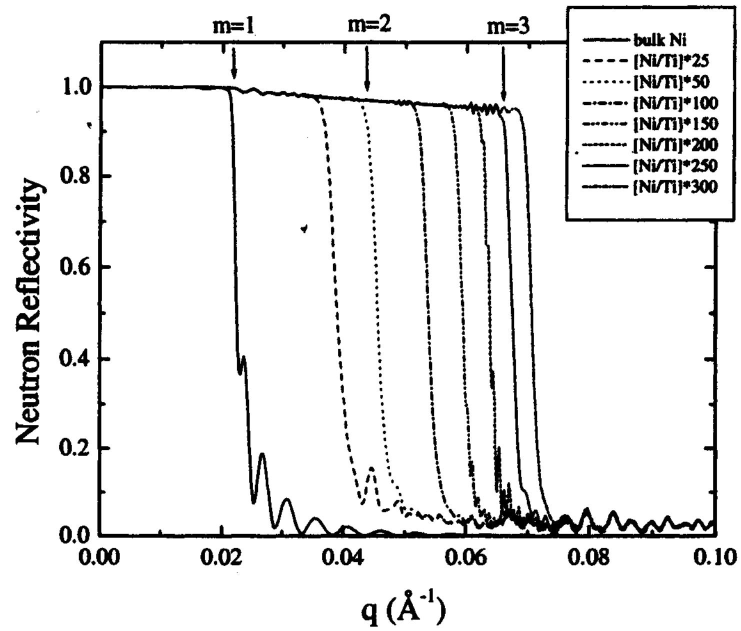

Fig. 2. Calculated neutron reflectivity of Ni/Ti supermirrors as a function of the number of bilayers.

In our example the correlation between number of bilayers and q_c of the resulting supermirrors is approximately given by

$$\text{Number of bilayers} \sim 3 \times m^4. \tag{4}$$

This function is plotted in Fig. 3. It is worthwhile to note that the exact relation always depends on the design reflectivity of the SM.

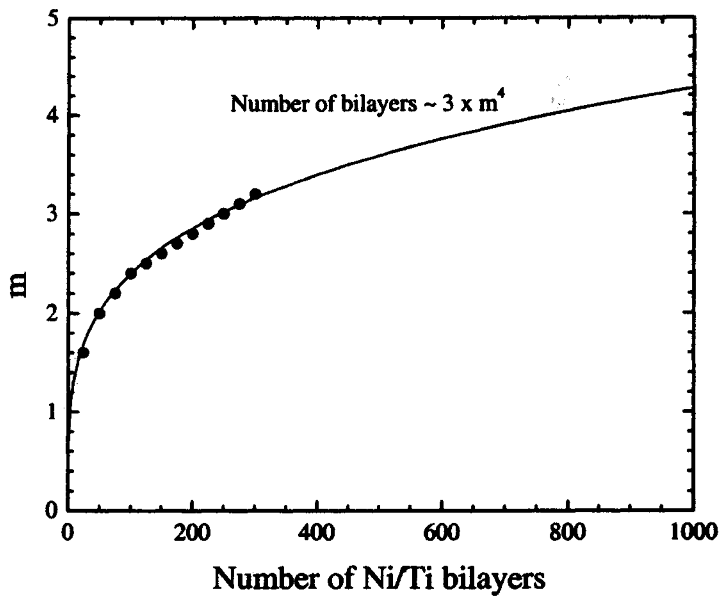

Fig. 3. Correlation between number of Ni/Ti bilayers and the m-values of the supermirrors.

Figure 3 also points out the requirement for adding more and more bilayers in order to reach very high critical q values. This approach is technically limited due to the following reasons:

i) With an increasing number of layers, quality correspondingly suffers due to the increasing amplification of interface roughness.
ii) Diffusion plays an increasing role especially for high-m supermirrors. The smallest single layer thickness of an m=4 supermirror is about 40 Å. For metallic multilayers, it is almost impossible to achieve rms-roughness values less than 5 Å.
iii) The technical demands and fabrication time needed for depositing high-m supermirrors is roughly proportional to the number of layers; therefore, the cost for high-performance supermirrors rises very steeply.
iv) The control of mechanical strain becomes more and more difficult for high-m supermirrors having very high total film thickness, e.g. approximately 35,000 Å in the case of m=3.5 mirrors. Associated with this is the danger of mechanical failure of the films (cracks or extensive peeling).

Therefore, it seems that m=4 should be considered as a practical limit for the m-value of supermirrors, at least with the deposition technology available today. In fact, m=4 supermirrors with 80% reflectivity at q_c have as yet only been produced on laboratory scale.[10] To the best of our knowledge, large area samples of these mirrors for actual applications have not reached more than 60% reflectivity.

4.2 Isotope substitution

Enhancement of the critical q-value of a supermirror and its reflectivity function may be achieved by artificially increasing the contrast in scattering length density, $\Delta_{N\cdot b}$, between the materials A,B constituting the mirror. Table 1 lists $\Delta_{N\cdot b}$ values for the cases in which natural Ni and Ti are substituted by more favorable isotopes or alloyed with other elements, for example hydrogen or carbon.

Material A	N·b (10^{-6} Å^{-2})		Material B	$\Delta_{N \cdot b}$ (10^{-6} Å^{-2})
Ni	9.4044	-1.945	Ti	11.3494
NiC	9.950	-1.945	Ti	11.8950
^{58}Ni	13.1479	-1.945	Ti	15.0929
^{58}Ni	13.1479	-3.4397	^{48}Ti	16.5876
^{58}Ni	13.1479	-6.0	TiH	19.1479
^{58}Ni	13.1479	-7.9435	^{62}Ni	21.0914

Tab. 1. Scattering length densities N·b of natural Ni and Ti, respectively, and possible isotope substitutions.

Figure 4 shows the effect of isotope substitution of the Ni layers for a Ni/Ti supermirror with 300 bilayers (in this case the refraction corrected Mezei formula has been used to calculate the layer sequence; absorption/incoherent scattering corrections have been included; the interfaces were assumed to be ideal). As can be seen, there are significant improvements in m-value and reflectivity for the ^{58}Ni/Ti supermirror compared to the same mirror made out of natural Ni and Ti. Further gain in using even more exotic combinations like ^{58}Ni/^{62}Ni is only a few percent in reflectivity.

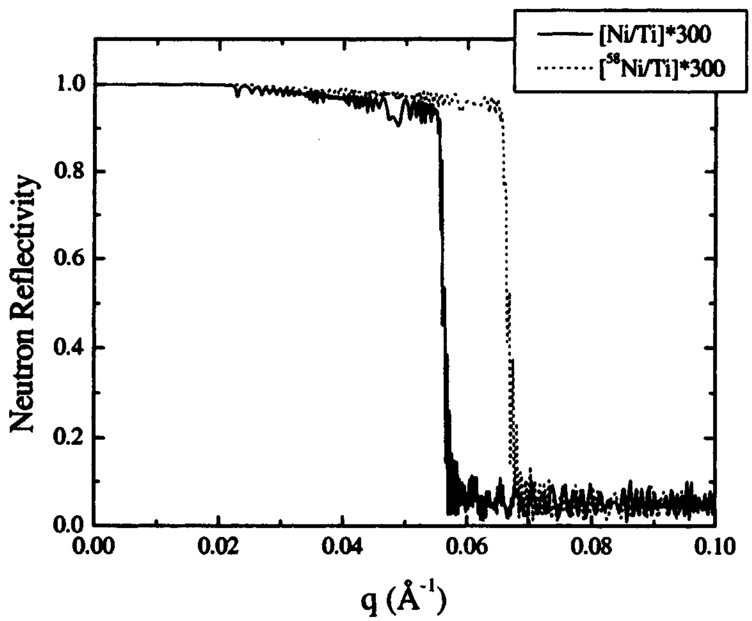

Fig. 4. Effect of isotope substitution in the Ni layers.

Despite the attractive possibilities of improving a supermirror's performance by using isotopes, this approach has very high impact on production costs; therefore it is unlikely that isotope substitution will play a major role in large-scale production of supermirrors.

4.3 Interface roughness and other imperfections

Supermirrors with 3.6 times the critical q of Ni became commercially available only recently after years of R&D at Paul Scherrer Institute (PSI) / Switzerland. A general drawback of high-m mirrors is that the reflectivity function of these coatings is far from being perfect (to a lesser extent this is also true for lower-m supermirrors, e.g. m=2 and m=3). In large-scale production of m=3.6 supermirrors, typical reflectivities of R=0.6-0.7 are reached at q_c. Theoretically, assuming a perfect layering, the reflectivity function should be considerably higher, on the order of 90% at q_c (absorption due to the enormous total thickness of approximately 35,000 Å and incoherent scattering are taken into account in the calculations). Obviously, large performance losses are caused by imperfections at the Ni/Ti interfaces and by the surface roughness of the substrate. So far, interface diffusion is thought to be the main reason for the low measured reflectivities; however, there might be other contributing factors that are not yet well investigated, for example small-angle scattering on the grain structure. Major distortions to the reflectivity may also result from limited coherence due to deviations from the design layer thicknesses, as was pointed out by Mezei.[7] It seems to be quite a challenge to keep the positions of the interfaces close to the nominal values in order to maintain coherent interference, particularly for supermirrors with very high m-values and the corresponding small individual layer thicknesses. For example, in the case of an m=3.5 supermirror, about 26 coherently reflecting bilayers are required for optimum reflectivity at q_c (where the individual layer thicknesses are about 40 Å). In order to satisfy the ± 45° phase difference criteria (cf. paragraph 3), offsets of actual positions of interfaces must be less than 10 Å.

There have been some suggestions recently to avoid amplification of interface roughness that occurs naturally when several hundred bilayers are being deposited. One idea is to smooth the layers after a certain fraction of the total deposition process. For example, Soyama et al. have applied ion polishing in combination with ion beam sputtering.[11] They achieved a decrease in the rms roughness of Ni films by ion-polishing from 6.5 Å to 3.5 Å.

5. PERFORMANCE GAINS FOR THE SNS NEUTRON REFLECTOMETER

This section demonstrates possible gains in instrument performance that may be achievable by improving high-m supermirror coatings. The proposed SNS Magnetism Reflectometer serves as an example. The basic layout of this instrument is illustrated in Fig. 5. Neutrons from the cold liquid hydrogen moderator are guided to the sample position at an 18 m distance via a combination of a channel beam bender and a tapered neutron guide. The bender (length: 5 m) is used to minimize high-energy neutron background at the sample position. It deflects the useful part of the wavelengths distribution (λ > 1.5 Å) by 2° horizontally and feeds it into a 9 m long focusing section, which compresses the beam size to match a typical sample size of 25 mm^2. High-energy neutrons cannot follow this curvature and are scattered and absorbed by appropriate shielding. Neutrons scattered by the sample will be counted by a two-dimensional multidetector at a 19 m distance from the moderator. The wavelength is determined by time-of-flight. The instrument is designed for 60 Hz operation, the normal source frequency of SNS. Bandwidth choppers restrict the total bandwidth of neutrons that are incident onto the sample to $\Delta\lambda$ = 3.5 Å. If, for example, the most intense wavelength band from 2.6 Å to 6.1 Å is used for data collection at the SNS instrument, a neutron flux of approximately 3.7 x 10^9 neutrons/cm^2/s (at guide exit) can actually be used for concurrent data collection.

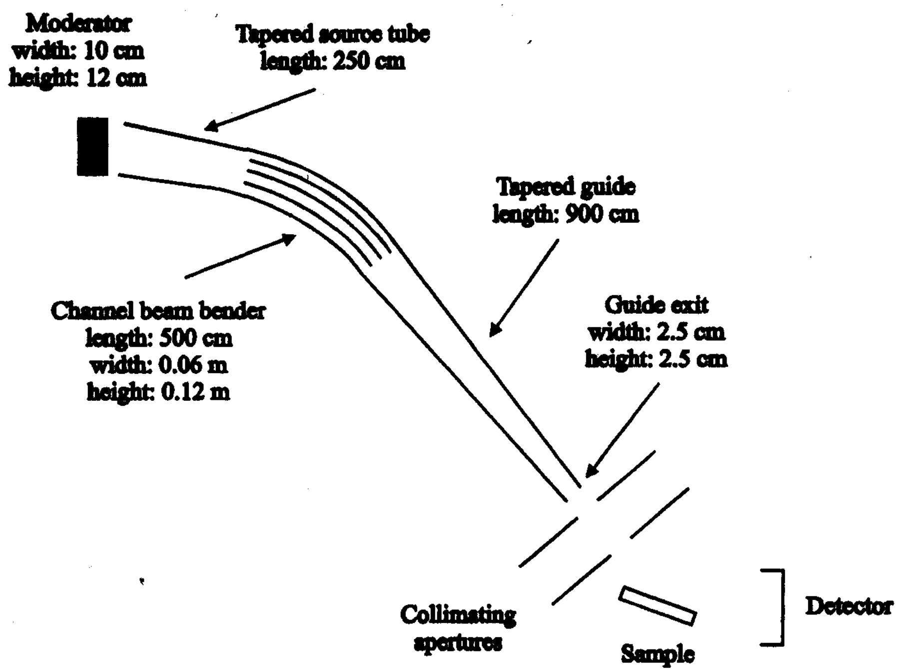

Fig. 5. Schematic layout of Magnetism Reflectometer to be built at SNS (top view).

The neutron guide system of the instrument has been optimized by Monte Carlo (MC) simulations using the program IDEAS.[12] The above stated flux number implies that m=3.5 supermirrors with 65% reflectivity at the critical edge will be utilized for all guide surfaces. This specification is challenging but does not seem to be beyond the capabilities of current guide vendors.

Figure 6 shows the effect of varying the reflectivity value (at q_c) for the above instrument configuration in the wavelength range up to 14 Å. In order to reflect a realistic situation in which large guide gains can be expected, we calculate flux on sample for a low-resolution experiment. In this case a highly divergent beam can be utilized. In particular, we assume: 25 mm x 25 mm sample size, 20° incident angle, and 10% angular resolution. The latter is achieved by using a pair of slits with 0.5 m distance from each other, which is located between the exit of the tapered guide and the sample position. The intensities displayed in Fig. 6 have been integrated over 5% wide neutron wavelength bins. Note that the sharp wavelength cut off at about 2 Å results from using the beam bender.

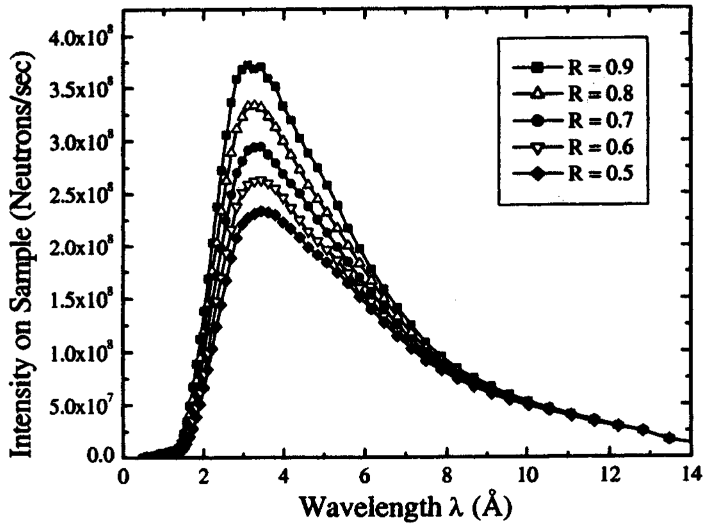

Fig. 6. Effect of different reflectivity (R) values (at q_c) for m = 3.5 supermirrors used as coating in the Magnetism Reflectometer neutron guide. The reflectivity function between q_c(Ni) and q_c(supermirror) was assumed to be linear.

Figure 7 shows the enhancement in flux-on-sample that may be achievable if supermirrors with higher reflectivity at q_c could be produced in large quantities. The intensity gain functions have been obtained by normalizing the flux values of Fig. 6 relative to the R=0.5 data. It can be seen from Fig. 7, that the short wavelength intensity in particular would be significantly increased.

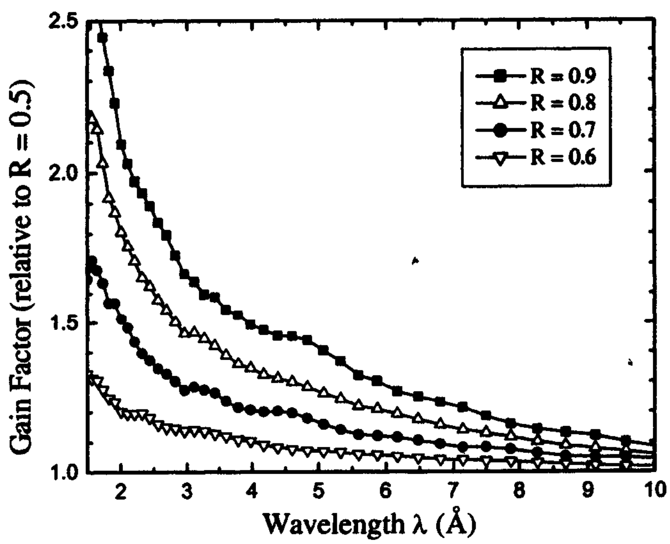

Fig. 7. Neutron intensity gain of various supermirror guide coatings (with different R-values at q_c) relative to a R=0.5 coating.

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